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Effect of carbon nanotube agglomeration on the piezoresistive response of polymer nanocomposite architected materials

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***Abstract*—**The addition of carbon nanotubes (CNTs) into a polymer matrix results in a nanocomposite material with piezoresistive behavior. This piezoresistive behavior has been exploited for strain sensing and self-sensing applications. Architected materials possess unusual properties, such as auxeticity, that makes them suitable for a wide range of applications, e.g., energy absorption, vibration mitigation, or sensors. This has resulted in an increased usage of nanocomposites to fabricate architected materials with added multifunctionality. However, one prevalent issue in the fabrication of nanocomposites is the agglomeration of nanofillers, e.g., CNTs. Usually, a homogeneous distribution of CNTs is desired to improve the piezoresistive response of nanocomposites. The presence of agglomerations may reduce the electrical conductivity of the nanocomposite, and as a result make it less sensitive to changes in electrical resistance. Because of this, a computational study on the effect of CNT agglomeration on the piezoresistive response of architected materials is presented. Two types of architected materials are studied, viz., re-entrant (auxetic) and hexagonal (non-auxetic) two-dimensional lattices. This is done to compare the piezoresistive response in these two scenarios and how agglomeration of CNTs affects each type of lattice. Then, in this study, a micromechanical model for electrical conductivity of polymers filled with CNTs is modified to account for changes in conductivity due to strain. Agglomeration is modeled in the micromechanical model through two agglomeration parameters that take values in the range [0, 1]. Different pairs of values for the agglomeration parameters are considered to analyze the effect of different CNT distributions. Values of the agglomeration parameters are carefully chosen as some pairs of values are physically unfeasible. Then, for a given pair of agglomeration parameters, electrical conductivity can be calculated as a function of strain at each point of an architected material when a voltage boundary condition is applied. Mechanical deformation is applied via a displacement boundary condition. The resistance of the architected material is then calculated at each strain increment to obtain the piezoresistive response and calculate the gauge factor. Such simulations are carried out via finite element simulations. Finally, the piezoresistive curves and gauge factors of auxetic and non-auxetic architected materials are compared in terms of the agglomeration parameters. Insights on how to tune the gauge factor and reduce undesired effects of CNT agglomeration are discussed based upon the results obtained from simulations.

***Keywords-component*—piezoresistivity; nanocomposites; carbon nanotube; architected materials**

I. Introduction

The addition of carbon nanotubes (CNTs) into a polymer matrix results in an electrically conductive nanocomposite [1], [2]. This is possible due to the CNTs forming a percolated network that allows the flow of electricity throughout the nanocomposite [3]. When strain is applied, some CNTs may be separated from the CNT network or may form new conductive paths due to the deformation of the percolated network [4]. CNT/polymer nanocomposites are studied for strain sensing applications by taking advantage of this piezoresistive effect [5], [6].

Architected metamaterials are those where a porous structure is fabricated using a base material [7]. The base material is placed in a strategic way such that the behavior of the porous structure depends on its architecture and not the composition of the base material [7], [8]. For instance, a lattice of struts forming hexagons exhibits the Poisson's effect, i.e., its cross-section gets reduced when subjected to tension. In turn, if struts are arranged forming re-entrant unit cells (concave hexagons), then the lattice exhibits auxeticity. An auxetic material increases its cross-sectional area when subjected to tension, i.e., they have a negative Poisson's ratio. Auxetic materials have potential to be used for energy absorption applications [9] and a sensors that can conform to curved surfaces [8]. By using a nanocomposite as base material, auxetic architected structures with sensing capabilities may be obtained.

One issue in the fabrication of nanocomposites is the agglomeration of CNTs, which results in the reduction in their electrical conductivity [2], [10]. On the other hand, CNTs may be segregated in certain regions of the nanocomposite, which usually results in more electrically conductive materials [11]. As the electrical conductivity is affected by CNT dispersion (segregation and agglomeration), the piezoresistive response of architected materials that use nanocomposites as the base material will also be affected. Thus, it is important to take in consideration the dispersion of CNTs when modeling nanocomposite architected structures. Because of this, in this paper the effect of agglomeration of CNTs in hexagonal and re-entrant lattices is studied. These lattices are chosen to observe the differences in piezoresistive response of auxetic and non-auxetic lattices. A micromechanics model that takes into consideration the agglomeration and segregation of CNTs is used for the electrical conductivity of the base material. This micromechanics model also obtains the electrical conductivity as a function of strain. Finite element (FE) simulations of tensile tests are performed on both types of lattices. The micromechanics model is used within the FE simulations to obtain the electrical resistance of the lattices as a function of strain, i.e., their piezoresistive response. Piezoresistive responses of the auxetic and non-auxetic lattices are compared for different degrees of agglomeration and segregation of CNTs.

II. Methodology

A. Design of architected materials

Two architected material topologies are considered, i.e., an hexagonal and a re-entrant lattice. The hexagonal lattice exhibits the Poisson effect while the re-entrant lattice exhibits auxeticity. Both of these lattices are studied in two dimensions (2D). Fig. 1 shows the parameters that define the geometry of the lattices herein studied. In Fig. 1a, $l_h = 8$ mm and $t_h = 4$ mm, which results in $H_h = 170.11$ mm and $W_h = 98.21$ mm. In Fig. 1b, $h_r = 10$ mm, $t_r = 2$ mm, $l_r = \sqrt{3}h_r/2$, and $\theta = 70°$, which results in $H_r = 130.16$ mm and $W_r = 117.26$ mm. These measurements result in lattices with similar volume fractions so that both lattices have a similar amount of base material. The hexagonal lattice has a volume fraction of 0.3978, while for the re-entrant lattice it is 0.3427.

B. Piezoresitive study of architected materials

FE simulations of the lattices shown in Fig. 1 are performed using COMSOL Multiphisics (v6.3, COMSOL AB, Stockholm, Sweden). To reduce computational cost, simulations are performed in 2D using triangular elements setting their size to 'Extra fine' within COMSOL. Tension tests are performed by setting the top edges of the lattices to a prescribed displacement and fixing the bottom boundary edges. At the same time, a voltage of 10 V is applied to the top edges while the bottom edges are set to ground.

The electrical conductivity of the lattice is calculated using the micromechanics model presented in [2]. This micromechanics model accounts for the agglomeration and segregation of CNTs via two agglomeration parameters: χ and ζ. In that model, the nanocomposite is considered to have a dispersed phase and an agglomerated phase. These phases are illustrated in Fig. 2. In the dispersed phase there may be percolation while in the agglomerated phase there is no percolation as the CNT agglomerates are assumed to not be in electrical contact with the dispersed phase. Then, χ is the fraction of volume in the nanocomposite that corresponds to the agglomerated phase. ζ is the fraction of CNTs that are present in the agglomerated phase. For instance, $\chi = 0.25$ means that the agglomerated phase corresponds to 25 % of the nanocomposite's volume. A value of $\zeta = 0.25$ means that 25 % of the CNTs are present in the agglomerated phase, while the rest is in the dispersed phase. Following [12], the micromechanics model in [2] is modified to obtain the electrical conductivity as a function of strain. Then, in the piezoresistivity model herein used, two parameters can be employed to control the piezoresistive response. The first parameter, C_1, is used to define a linear increase of the average CNT separation as

$$d_a = d_{a,0}(1 + C_1\varepsilon_{eq}). \tag{1}$$

Here, $d_{a,0}$ is the average CNT separation at zero strain and ε_{eq} is an equivalent strain. Similarly, the parameter C_2 is used to linearly increase the height barrier, λ_0, as

$$\lambda = \lambda_0(1 + C_2\varepsilon_{eq}). \tag{2}$$

Due to performing tensile tests where the prescribed displacement is along the positive y-direction, we use $\varepsilon_{eq} = \varepsilon_{yy}$. The piezoresistive model is implemented in Matlab (R2024b, The MathWorks Inc., Natick, MA, USA) and used within COMSOL to obtain the electrical conductivity of the lattices. The electrical resistance of the lattices is obtained from COMSOL at various strain values.

Two parameter studies are performed to evaluate the piezoresistive response of the lattices in Fig. 1. The piezoresistive curve and the gauge factor are obtained for each simulation performed. The piezoresistive curve is the relative change in electrical resistance, $(R - R_0)/R_0$, as a function of strain. Here R_0 is the electrical resistance of the lattice at zero strain and R is the electrical resistance as a function of strain. The gauge factor, k, is the slope of the piezoresistive curve. In the first parameter study, multiple pairs of values for χ and ζ are simulated while using $C_1 = 5$ and $C_2 = 1.5$ [12]. The values of the agglomeration parameters are chosen so that they do not fall within physically unfeasible regions [2]. Initially, ζ is set to zero and the value of χ is increased from 0 to 0.75. This corresponds to a segregated state due to having all CNTs in a specific and smaller volume of the nanocomposite. As a result, the dispersed phase has a higher CNT concentration and becomes more electrically conductive. Then, χ is set to 0.25 and the value of ζ is increased from 0 to 0.75. This corresponds to starting from a segregated (and highly conductive) nanocomposite to one with increased CNT agglomerates as ζ increases. In the second parameter study, both agglomeration parameters are set to zero and the piezoresistive parameters are varied. Having $\chi = \zeta = 0$ results in a nanocomposite with a homogeneous distribution of CNTs.

The material parameters for the piezoresistive model are shown in Table I. All parameters in that table, except for CNT volume fraction, correspond to those in [2] for CNT/polylactic

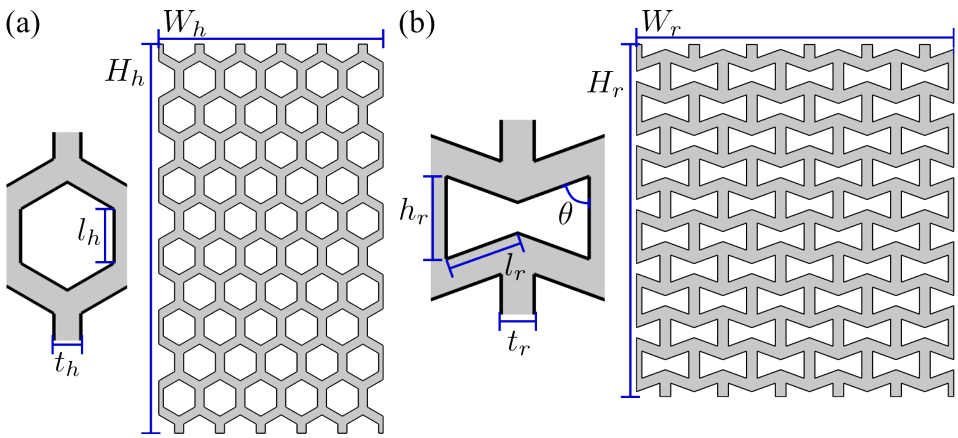

Figure. 1. Architected structures and dimensions. (a) Hexagonal lattice, (b) re-entrant lattice.

TABLE. I
PARAMETERS FOR PIEZORESISTIVE MODEL.

Parameter	Value
CNT length [μm]	30
CNT radius [nm]	5
CNT volume fraction	0.03
Percolation threshold	3.239×10^{-3}
Cutoff for tunneling [nm]	1.8
Height barrier (λ_0) [eV]	1.5
Conductivity of polymer [S/cm]	10^{-16}
Conductivity of CNT [S/cm]	9.87

acid nanocomposites. A CNT volume fraction of 0.3 is above percolation in that work, and thus can show piezoresistivity. Material parameters for the FE simulations are shown in Table II.

III. Results and Discussion

A. Study on agglomeration parameters

The piezoresisitive curves for lattices with homogeneous and segregated states are shown in Fig. 3. For the lattices with homogeneous distributions of CNTs ($\chi = \zeta = 0$), the piezoresistive response is largely different. The hexagonal lattice is more sensitive to changes in strain as the change in electrical resistance due to the same deformation is larger. In both cases, as the nanocomposite becomes more segregated (i.e., as χ increases), the change in electrical is more pronounced. Here, an increase in χ with $\zeta = 0$ results in having no CNTs in the agglomerated phase. Then, all CNTs are present in the dispersed phase that gets smaller within the nanocomposite base material as χ increases. That is, CNTs become segregated in the dispersed phase which occupies a fraction of the lattice equal to $1-\chi$. It is also observed from Fig. 3 that the larger the segregation the larger the sensitivity to strain. This is reflected in the gauge factors (slope of the piezoresistive curves), which are shown in Table III. In both lattices, when increasing χ from 0 to 0.25, there is a small increment of the gauge factor (below 10%). In contrast, when increasing χ from 0.50 to 0.75, the gauge factor of the hexagonal lattice increases around 50% and the one for the re-entrant lattice more than doubles.

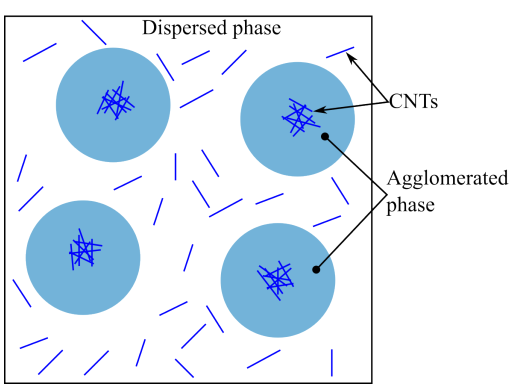

Figure. 2. Schematic representation of the dispersed and agglomerated phases of a nanocomposite.

Fig. 4 shows the piezoresistive curves for the agglomerated states. In all those curves, χ is set to 0.25 and ζ increases from 0 to 0.75. This means that as ζ increases, more CNTs are present in the agglomerated phase and less are present in the dispersed phase. This results in a reduction of the electrical conductivity of the dispersed phase and so also of the lattices. For both lattices, negligible changes were observed when $0 < \zeta < 0.25$ and, thus, curves obtained with such values are not shown. When ζ reached a value of 0.25, i.e., the same of χ, changes were observed in the piezoresistive curves of both lattices. This suggests that $\zeta = \chi$ is a threshold for non-negligible effects of agglomeration in the piezoresistive response of the lattices herein studied. From Fig. 4, when

TABLE. II
PARAMETERS FOR FE SIMULATIONS.

Parameter	Value
Polymer density [g/cm^3] [13]	1.25
Young's modulus [GPa] [13]	3.5
Poisson's ratio [14]	0.35
Relative permitivity [15]	3.0

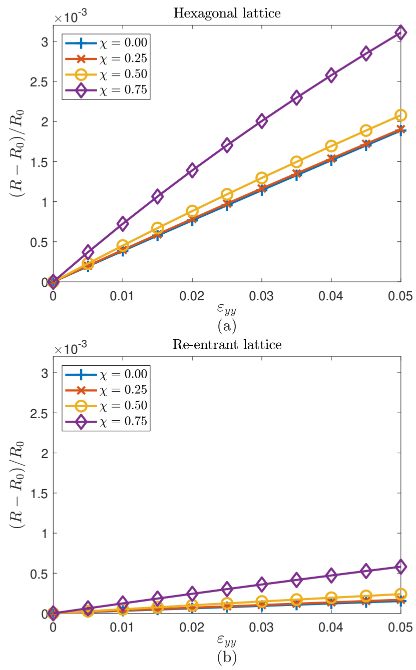

Figure. 3. Piezoresistive curves for lattices with a homogeneous distribution of CNTs and segregated states ($\zeta = 0$). (a) Hexagonal lattice, (b) re-entrant lattice.

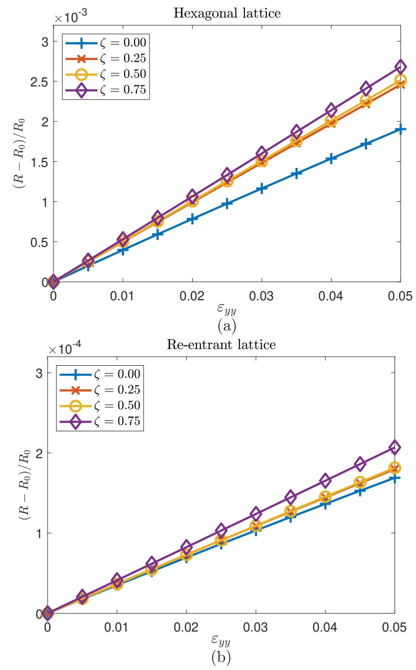

Figure. 4. Piezoresistive curves for lattices with varying degrees of CNT agglomeration ($\chi = 0.25$). (a) Hexagonal lattice, (b) re-entrant lattice.

$\zeta \geq \chi$, CNT agglomeration makes the lattices more sensitive to changes in strain. As observed in Fig. 3, re-entrant lattices are less sensitive to strain compared to hexagonal lattices. Note that the limit of the y-axis in Fig. 4b is 10% of that in Fig. 4a. Gauge factors for the plots in Fig. 4 are shown in Table IV. For both types of lattices, an increase in CNT agglomeration (via an increase of ζ) results in an increase of gauge factor. From both the curves in Fig. 4 and the gauge factors in Table IV, it is observed that the largest change in piezoresistive behavior for hexagonal lattices happened when increasing ζ from 0 to 0.25. In contrast, the largest change in piezoresistive behavior for re-entrant lattices was observed when increasing ζ from 0.50 to 0.75. These results suggest that the re-entrant lattice is more sensitive to changes in strain for large degrees of CNT agglomeration while the hexagonal lattice is more sensitive for low degrees of agglomeration. It is also observed that the changes in piezoresistive response due to agglomeration are lower compared to the changes obtained through CNT segregation.

B. *Study on piezoresistive parameters*

Fig. 5a shows the piezoresistive curves for hexagonal lattices and homogeneous distribution of CNTs ($\chi = \zeta = 0$). In all those curves, $C_1 = 5.0$ while C_2 is increased from 1.5 to 10. It is clear that the effect of C_2 is negligible, even when increasing it value nearly ten times. From (2), a larger value of C_2 results in a larger increase of λ due to strain. Then, the negligible effect of C_2 is a direct consequence of the small effect of increasing λ in the electrical conductivity of CNT nanocomposites [10].

Piezoresistive curves for hexagonal lattices and homogeneous distribution of CNTs are shown in Fig. 5b, where $C_2 = 1.5$ for all curves. It is clear that there is an increase in the piezoresistive response by increasing C_1. This is a direct consequence of the large effect of d_a on the electrical conductivity of CNT nanocomposites as it defines the conductivity of the equivalent filler in micromechanics models [2], [10]. Then, although the micromechanics piezoresistive model requires two parameters to define the piezoresistive response, curves in Fig. 5 suggest that only one is needed. Thus, such micromechanics models (e.g., [12]) could be turned into a

TABLE. III
GAUGE FACTORS FOR HEXAGONAL AND RE-ENTRANT LATTICES WITH SEGREGATION OF CNTS ($\zeta = 0$).

χ	Hexagonal	Re-entrant
0.00	0.0377	0.0031
0.25	0.0381	0.0034
0.50	0.0416	0.0048
0.75	0.0622	0.0116

TABLE. IV
GAUGE FACTORS FOR HEXAGONAL AND RE-ENTRANT LATTICES WITH AGGLOMERATION OF CNTS ($\chi = 0.25$).

ζ	Hexagonal	Re-entrant
0.00	0.0381	3.3797×10^{-3}
0.25	0.0492	3.5985×10^{-3}
0.50	0.0503	3.6301×10^{-3}
0.75	0.0536	4.1378×10^{-3}

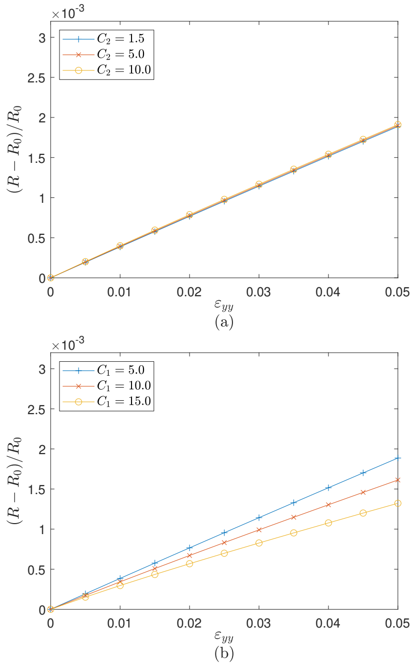

Figure. 5. Piezoresistive curves for a hexagonal lattice with homogeneous distribution of CNTs. (a) $C_1 = 5.0$, (b) $C_2 = 1.5$.

single piezoresistive parameter models or other piezoresistive parameters may be introduced. For instance, a dependence of the CNT efficiency, η, on strain may be introduced as

$$\eta = \eta_0(1 + C_3\varepsilon_{eq}). \tag{3}$$

where η_0 is the CNT efficiency at zero strain. CNT efficiency is the fraction of CNTs that actually percolate and, thus, carry electrical current [16].

IV. Conclusions

It was found that hexagonal (non-auxetic) lattices were more sensitive to changes in strain compared to re-entrant (auxetic) lattices. Dispersion of CNTs plays in important role in the piezoresistive response of the architected materials herein studied. Segregation of CNTs resulted in an increase in sensitivity to strain in both lattices. CNT agglomeration also increased sensitivity to strain in both lattices but to a lesser extent compared to segregation. Re-entrant lattices were more sensitive to changes in strain for large CNT agglomeration while hexagonal lattices were more sensitive for low CNT agglomeration. Thus, promoting segregation in architected structures may result in sensors with improved strain sensitivity.

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